Breakdown of the few-level approximation in collective systems

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The validity of the few-level approximation in dipole-dipole interacting collective systems is discussed. As example system, we study the archetype case of two dipole-dipole interacting atoms, each modelled by two complete sets of angular momentum multiplets. We prove that the dipole-dipole induced energy shifts between collective two-atom states depend on the length of the vector connecting the atoms, but not on its orientation, if complete multiplets are considered. For this, a strong link between any two alignments with a fixed distance between the atoms is established. The simplification of the atomic level scheme by artificially omitting Zeeman sublevels in a few-level approximation, however, generally leads to incorrect predictions.

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The theoretical analysis of any non-trivial physical problem typically requires the use of approximations. A key approximation facilitated in most areas of physics reduces the complete configuration space of the system of interest to a smaller set of relevant system states. In the theoretical description of atom-field interactions, the essential state approximation entails neglecting most of the bound and continuum atomic states [1, 2, 3]. The seminal Jaynes-Cummings-Model [4] takes this reduction to the extreme in that only two atomic states are retained. Obviously, it is essential to in detail explore the validity range of this reduction of the configuration space. The few-level approximation usually leads to theoretical predictions that are well verified experimentally [1, 2], and is generally considered as understood for single-atom systems. It fails, however, to reproduce results of quantum electrodynamics [5]. The situation becomes even less clear in collective systems, where the individual constituents interact via the dipole-dipole interaction, despite the relevance of collectivity to many areas of physics. Examples for such systems can be found in ultracold quantum gases [6, 7, 8], trapped atoms [9, 10, 11], or solid state systems [12, 13], with applications, e.g., in quantum information theory [14, 15, 16, 17].

Therefore in this Letter, we discuss the validity of the few-level approximation in dipole-dipole interacting collective systems. For this, we study the archetype case of two dipole-dipole interacting atoms, see Fig. 1(a). Experiments of this type have become possible recently [9, 10, 12]. Each atom is modelled by complete sets of angular momentum multiplets, as shown in Fig. 1(b). We find that the few-level approximation in general leads to unphysical predictions if it is applied to the magnetic sublevels of this system. In particular, we prove that the dipole-dipole induced energy shifts between collective two-atom states are invariant under rotations of the separation vector $\boldsymbol{R}$ if complete multiplets are considered. Throughout this analysis, we derive a strong link between any two orientations of $\boldsymbol{R}$ with a fixed distance between the atoms. The artificial omission of any of the Zeeman sublevels of a multiplet leads to a spurious dependence of the energy shifts on the orientation, and thus to incorrect predictions. For example, if only one excited state $|e\rangle$ and the ground state $|g\rangle$ are retained, we recover the position-dependent energy splitting between the entangled two-particle states $(|e,g\rangle \pm |g,e\rangle)/\sqrt{2}$ that has previously been reported for a pair of two-level systems [2, 3]. Our results can be generalized to more complex angular momentum multiplets.

We describe each atom by a $S_0 \leftrightarrow P_1$ transition shown in Fig. 1(b) that can be found, e.g., in ^{40}Ca atoms. We choose the z axis as the quantization axis, which is distinguished by an external magnetic field that induces a Zeeman splitting δ of the excited states. The orientation of $\boldsymbol{R}$ is defined relative to this quantization axis. We begin with the introduction of the master equation which governs the atomic evolution of the system shown in Fig. 1. The internal state $|i_\mu\rangle$ of atom μ is an eigenstate of $J_z^{(\mu)}$, where $\boldsymbol{J}^{(\mu)}$ is the angular momentum operator of atom μ ($\mu \in \{1,2\}$). In particular, the P_1 multiplet with $J=1$ corresponds to the excited states $|1_\mu\rangle$, $|2_\mu\rangle$ and $|3_\mu\rangle$ with magnetic quantum numbers $m=-1$, 0 and 1, respectively, and the S_0 state is the ground state $|4_\mu\rangle$ with $J=m=0$. The raising and lowering operators on the

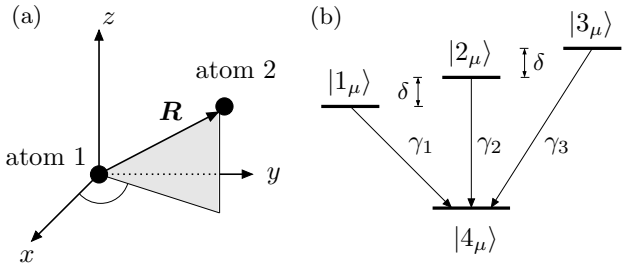

FIG. 1: (a) The system of interest is comprised of two identical atoms that are located at $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$, respectively. $\boldsymbol{R}=\boldsymbol{r}_2-\boldsymbol{r}_1$ is the relative position of atom 2 with respect to atom 1. (b) Level structure of atom $\mu \in \{1,2\}$ which we employ to illustrate our results. The ground state is a S_0 singlet state, and the three excited levels are Zeeman sublevels of a P_1 triplet. δ is the frequency splitting of the upper levels.

$|4_\mu\rangle \leftrightarrow |i_\mu\rangle$ transition of atom μ are ($i \in \{1,2,3\}$)

$$S_{i+}^{(\mu)} = |i_\mu\rangle\langle 4_\mu| \quad \text{and} \quad S_{i-}^{(\mu)} = |4_\mu\rangle\langle i_\mu| \,. \tag{1}$$

The total system Hamiltonian for the two atoms and the radiation field is $H = H_\mathrm{A} + H_\mathrm{F} + V$, where

$$\begin{aligned} H_\mathrm{A} =& \hbar \sum_{i=1}^{3}\sum_{\mu=1}^{2} \omega_i \, S_{i+}^{(\mu)} S_{i-}^{(\mu)} \,, \quad H_\mathrm{F} = \sum_{\boldsymbol{ks}} \hbar\omega_k a_{\boldsymbol{ks}}^\dagger a_{\boldsymbol{ks}} \,, \\ V =& -\hat{\boldsymbol{d}}^{(1)} \cdot \hat{\boldsymbol{E}}(\boldsymbol{r}_1) - \hat{\boldsymbol{d}}^{(2)} \cdot \hat{\boldsymbol{E}}(\boldsymbol{r}_2) \,. \end{aligned} \tag{2}$$

In these equations, H_A describes the free evolution of the two identical atoms, $\hbar\omega_i$ is the energy of state $|i_\mu\rangle$ and we choose $\hbar\omega_4 = 0$. H_F is the Hamiltonian of the vacuum field and V describes the interaction of the atom with the vacuum modes in dipole approximation. The electric field operator $\hat{\boldsymbol{E}}$ is defined as

$$\hat{\boldsymbol{E}}(\boldsymbol{r}) = i \sum_{\boldsymbol{ks}} \sqrt{\frac{\hbar\omega_k}{2\varepsilon_0 v}} \boldsymbol{\epsilon}_{\boldsymbol{ks}} e^{i\boldsymbol{k}\cdot\boldsymbol{r}} a_{\boldsymbol{ks}} + \mathrm{H.c.} \,, \tag{3}$$

where $a_{\boldsymbol{ks}}$ ($a_{\boldsymbol{ks}}^\dagger$) are the annihilation (creation) operators that correspond to a field mode with wave vector $\boldsymbol{k}$, polarization $\boldsymbol{\epsilon}_{\boldsymbol{ks}}$ and frequency ω_k, and v denotes the quantization volume. The electric-dipole moment operator of atom μ is a vector operator with respect to the angular momentum operator $\boldsymbol{J}^{(\mu)}$ of atom μ and reads

$$\hat{\boldsymbol{d}}^{(\mu)} = \sum_{i=1}^{3} \left[\boldsymbol{d}_i S_{i+}^{(\mu)} + \mathrm{H.c.} \right] . \tag{4}$$

We determine the dipole moments $\boldsymbol{d}_i = \langle i|\hat{\boldsymbol{d}}|4\rangle$ via the Wigner-Eckart theorem [18] and find $\boldsymbol{d}_1 = \mathcal{D}\,\boldsymbol{\epsilon}^{(+)}$, $\boldsymbol{d}_2 = \mathcal{D}\,\boldsymbol{e}_z$ and $\boldsymbol{d}_3 = -\mathcal{D}\,\boldsymbol{\epsilon}^{(-)}$, where $\mathcal{D}$ is the reduced dipole matrix element and the circular polarization vectors are $\boldsymbol{\epsilon}^{(\pm)} = (\boldsymbol{e}_x \pm i\boldsymbol{e}_y)/\sqrt{2}$.

Following the standard derivation of a master equation [1, 2, 3], we assume that the radiation field is initially in the vacuum state denoted by ϱ_F. Furthermore, we suppose that the total density operator factorizes into a product of ϱ_F and the atomic density operator ϱ at $t = 0$. The master equation for the reduced atomic density operator in Born approximation then takes the form

$$\begin{aligned} \partial_t \varrho =& -\frac{i}{\hbar}[H_\mathrm{A}, \varrho] \\ & - \frac{1}{\hbar^2} \int_0^t d\tau \mathrm{Tr}_\mathrm{F}\Big(\big[V, U(\tau)\big[V, \varrho_\mathrm{F}\varrho(t-\tau)\big]U^\dagger(\tau)\big] \Big) \,, \end{aligned} \tag{5}$$

where $U(\tau) = \exp[-i(H_\mathrm{A} + H_\mathrm{F})\tau/\hbar]$ and $\mathrm{Tr}_\mathrm{F}()$ denotes the trace over the vacuum modes. We evaluate the integral in Eq. (5) in Markov-approximation [1] and ignore all terms associated with the Lamb shift of the atomic levels. In addition, we employ the rotating-wave approximation and neglect anti resonant terms that are proportional to $S_{i+}^{(\mu)} S_{j+}^{(\nu)}$ and $S_{i-}^{(\mu)} S_{j-}^{(\nu)}$. We finally obtain

$$\partial_t \varrho = -\frac{i}{\hbar}[H_\mathrm{A}, \varrho] - \frac{i}{\hbar}[H_\Omega, \varrho] + \mathcal{L}_\gamma \varrho \,. \tag{6}$$

In this equation, the Hamiltonian H_Ω describes the coherent part of the dipole-dipole interaction and reads

$$H_\Omega = -\hbar \sum_{i,j=1}^{3} \Big\{ \Omega_{ij} S_{i+}^{(2)} S_{j-}^{(1)} + \mathrm{H.c.} \Big\} \,. \tag{7}$$

The coefficients Ω_{ij} are defined as [19, 20]

$$\Omega_{ij} = \frac{1}{\hbar} \left[\boldsymbol{d}_i^\mathrm{T} \overleftrightarrow{\chi}_\mathrm{re}(\boldsymbol{R})\, \boldsymbol{d}_j^* \right] , \tag{8}$$

and the tensor $\overleftrightarrow{\chi}_\mathrm{re}$ is the real part of the tensor $\overleftrightarrow{\chi}$ whose components $\overleftrightarrow{\chi}_{kl}$ for $k, l \in \{1,2,3\}$ are given by

$$\overleftrightarrow{\chi}_{kl}(\boldsymbol{R}) = \frac{k_0^3}{4\pi\varepsilon_0} \left[g_1(\eta)\,\delta_{kl} - g_2(\eta)\,\frac{\boldsymbol{R}_k \boldsymbol{R}_l}{R^2} \right] e^{i\eta} \,. \tag{9}$$

Here the vector $\boldsymbol{R}$ denotes the relative coordinates of atom 2 with respect to atom 1 [see Fig. 1(a)], $\eta = k_0 R$ and $g_1 = (\eta^{-1} + i\eta^{-2} - \eta^{-3})$, $g_2 = (\eta^{-1} + 3i\eta^{-2} - 3\eta^{-3})$. In the derivation of Eq. (9), the three transition frequencies ω_1, ω_2 and ω_3 have been approximated by their mean value $\omega_0 = ck_0$ (c: speed of light) [21]. This is justified since the Zeeman splitting δ is much smaller than the optical transition frequencies ω_i. The last term in Eq. (6) accounts for spontaneous emission and reads

$$\begin{aligned} \mathcal{L}_\gamma \varrho =& -\sum_{\mu=1}^{2}\sum_{i=1}^{3} \gamma_i \Big(S_{i+}^{(\mu)} S_{i-}^{(\mu)} \varrho + \varrho S_{i+}^{(\mu)} S_{i-}^{(\mu)} - 2 S_{i-}^{(\mu)} \varrho S_{i+}^{(\mu)} \Big) \\ & - \sum_{\substack{\mu,\nu=1 \\ \mu\neq\nu}}^{2} \sum_{i,j=1}^{3} \Gamma_{ij} \Big(S_{i+}^{(\mu)} S_{j-}^{(\nu)} \varrho + \varrho S_{i+}^{(\mu)} S_{j-}^{(\nu)} - 2 S_{j-}^{(\nu)} \varrho S_{i+}^{(\mu)} \Big) \,. \end{aligned} \tag{10}$$

The total decay rate of the excited state $|i\rangle$ of each of the atoms is given by $2\gamma_i$, where $\gamma_i = |\boldsymbol{d}_i|^2 \omega_0^3/(6\pi\epsilon_0\hbar c^3) = \gamma$ and we again employed the approximation $\omega_i \approx \omega_0$. The collective decay rates Γ_{ij} result from the vacuum-mediated dipole-dipole coupling between the two atoms and are determined by

$$\Gamma_{ij} = \frac{1}{\hbar} \left[\boldsymbol{d}_i^\mathrm{T} \overleftrightarrow{\chi}_\mathrm{im}(\boldsymbol{R})\, \boldsymbol{d}_j^* \right] , \tag{11}$$

where $\overleftrightarrow{\chi}_\mathrm{im} = \mathrm{Im}\overleftrightarrow{\chi}$ is the imaginary part of the tensor $\overleftrightarrow{\chi}$.

We now derive a general statement about the behavior of the master equation (6) under rotations of the separation vector $\boldsymbol{R}$. This will provide the theoretical foundation for our central results and physical interpretations that follow after the formal proof of the statement.

In addition to a given relative position $\boldsymbol{R}$ of the two atoms we consider a different geometrical setup where

the separation vector $\boldsymbol{P}$ is obtained from $\boldsymbol{R}$ by a rotation, $\boldsymbol{P} = \mathcal{R}_{\boldsymbol{u}}(\alpha)\boldsymbol{R}$. Here, $\mathcal{R}_u(\alpha)$ is an orthogonal 3×3 matrix that describes a rotation in the three-dimensional real vector space $\mathbb{R}^3$ around the axis $\boldsymbol{u}$ by an angle α. We show that there exists a unitary operator W such that

$$H_\Omega(\boldsymbol{P}) = W H_\Omega(\boldsymbol{R}) W^\dagger\,, \tag{12a}$$

$$\mathcal{L}_\gamma(\boldsymbol{P})\varrho = W\left[\mathcal{L}_\gamma(\boldsymbol{R}) W^\dagger \varrho W\right] W^\dagger\,, \tag{12b}$$

where $W = W_{\boldsymbol{u}}(\alpha)$ is given by

$$W_{\boldsymbol{u}}(\alpha) = \exp[-i\alpha\, \boldsymbol{J}^{(1)}\cdot\boldsymbol{u}/\hbar]\exp[-i\alpha\, \boldsymbol{J}^{(2)}\cdot\boldsymbol{u}/\hbar]\,. \tag{13}$$

Here the operator $\exp[-i\alpha\, \boldsymbol{J}^{(\mu)}\cdot\boldsymbol{u}/\hbar]$ describes a rotation around the axis $\boldsymbol{u}$ by an angle α in the state space of atom μ. The notation $H_\Omega(\boldsymbol{R})$ and $\mathcal{L}_\gamma(\boldsymbol{R})$ means that the coupling constants and collective decay rates in Eqs. (7) and (10) have to be evaluated at $\boldsymbol{R}$.

We proceed with the proof of Eq. (12). In a first step, we introduce the auxiliary operator $A_{\boldsymbol{R}} = W V_{\boldsymbol{R}} W^\dagger$, where $V_{\boldsymbol{R}}$ is the interaction Hamiltonian for a relative position of the atoms given by $\boldsymbol{R}$, and $W = W_{\boldsymbol{u}}(\alpha)$ is defined in Eq. (13). The evaluation of $A_{\boldsymbol{R}}$ involves only the transformation of the dipole operator of each atom. Since the matrix elements of vector operators transform like classical vectors under rotations (see, e.g., Sec. 3.10. in [18]), we find

$$W\, \hat{\boldsymbol{d}}^{(\mu)}\, W^\dagger = \sum_{i=1}^{3}\left[\tilde{\boldsymbol{d}}_i\, S_{i+}^{(\mu)} + \text{H.c.}\right], \tag{14}$$

where $\tilde{\boldsymbol{d}}_i = \mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)\boldsymbol{d}_i$. This shows that the only difference between the auxiliary operator $A_{\boldsymbol{R}}$ and $V_{\boldsymbol{R}}$ is that the dipole moments of the former are determined by $\tilde{\boldsymbol{d}}_i$ instead of $\boldsymbol{d}_i$. In a second step, we employ the tensor properties of $\overleftrightarrow{\chi}$ to find the following expression for the parameters $\Omega_{ij}(\boldsymbol{P})$ and $\Gamma_{ij}(\boldsymbol{P})$,

$$\hbar\Omega_{ij}(\boldsymbol{P}) = \left[\mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)\boldsymbol{d}_i\right]^{\mathrm{T}} \overleftrightarrow{\chi}_{\mathrm{re}}(\boldsymbol{R})\left[\mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)\boldsymbol{d}_j^*\right], \tag{15}$$

$$\hbar\Gamma_{ij}(\boldsymbol{P}) = \left[\mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)\boldsymbol{d}_i\right]^{\mathrm{T}} \overleftrightarrow{\chi}_{\mathrm{im}}(\boldsymbol{R})\left[\mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)\boldsymbol{d}_j^*\right]. \tag{16}$$

This important result shows that a rotation of the dipole moments $\boldsymbol{d}_i$ by $\mathcal{R}_{\boldsymbol{u}}^{-1}(\alpha)$ is formally equivalent to a rotation of $\boldsymbol{R}$ by $\mathcal{R}_{\boldsymbol{u}}(\alpha)$ in the master equation (6). From the combination of the results obtained in step one and two, we conclude that the exchange of $V_{\boldsymbol{R}}$ by $A_{\boldsymbol{R}}$ in the integral of Eq. (5) is equivalent to a rotation of the separation vector from $\boldsymbol{R}$ to $\boldsymbol{P} = \mathcal{R}_{\boldsymbol{u}}(\alpha)\boldsymbol{R}$,

$$I = \frac{-1}{\hbar^2}\int_0^t d\tau \mathrm{Tr}_{\mathrm{F}}\Big(\big[A_{\boldsymbol{R}}, U(\tau)\big[A_{\boldsymbol{R}}, \varrho_{\mathrm{F}}\varrho(\hat{\tau})\big]U^\dagger(\tau)\big]\Big) \tag{17}$$

$$= -\frac{i}{\hbar}[H_\Omega(\boldsymbol{P}),\varrho] + \mathcal{L}_\gamma(\boldsymbol{P})\varrho\,, \tag{18}$$

where $\hat{\tau} = t - \tau$. Note that the equality of Eqs. (17) and (18) holds under the same assumptions that led from Eqs. (5) to (6). In the second part of the proof we evaluate the integral in Eq. (17) in a different way. In the discussion following Eq. (9), we justified that $\mathcal{L}_\gamma$ and H_Ω depend only on the mean transition frequency ω_0. Here we employ exactly the same approximation [21] and replace the frequencies ω_i appearing in Eq. (17) by ω_0. In this case, $\boldsymbol{J}^{(\mu)}$ commutes with H_{A} such that the operators W and U commute, and the argument of the trace in Eq. (17) can be written as

$$W\big[V_{\boldsymbol{R}}, U(\tau)\big[V_{\boldsymbol{R}}, \varrho_{\mathrm{F}}\tilde{\varrho}(\hat{\tau})\big]U^\dagger(\tau)\big]W^\dagger\,, \tag{19}$$

where $\tilde{\varrho} = W^\dagger\varrho W$. In contrast to Eq. (17), the double commutator contains now the original interaction Hamiltonian $V_{\boldsymbol{R}}$ that corresponds to a setting with separation vector $\boldsymbol{R}$. We thus obtain

$$I = -\frac{i}{\hbar}[W H_\Omega(\boldsymbol{R}) W^\dagger, \varrho] + W\left[\mathcal{L}_\gamma(\boldsymbol{R}) W^\dagger\varrho W\right] W^\dagger\,. \tag{20}$$

Finally, the comparison of Eq. (20) with Eq. (18) establishes Eq. (12) which concludes the proof.

We now turn to the discussion of Eq. (12), which will lead to our central results. The Hamiltonian H_Ω describes the coherent part of the dipole-dipole interaction between the atoms. We employ Eq. (12a) to show that the eigenvalues of H_Ω depend only on the interatomic distance, but not on the orientation of the separation vector $\boldsymbol{R}$. At the same time, we present symbolic expressions for the eigenvalues and eigenstates of H_Ω. We find that H_Ω can be represented as

$$H_\Omega = \sum_{i,j=1}^{3}\left(\,[H_\Omega]_{ij}^{\mathcal{S}}\,|s_i\rangle\langle s_j| + [H_\Omega]_{ij}^{\mathcal{A}}\,|a_i\rangle\langle a_j|\,\right), \tag{21}$$

where the symmetric and antisymmetric states are defined as $|s_i\rangle = [\,|i,4\rangle + |4,i\rangle\,]/\sqrt{2}$ and $|a_i\rangle = [\,|i,4\rangle - |4,i\rangle\,]/\sqrt{2}$, respectively ($|i,j\rangle = |i_1\rangle\otimes|j_2\rangle$). The matrix elements of H_Ω in the subspace $\mathcal{S}$ spanned by the symmetric states $\{|s_1\rangle, |s_2\rangle, |s_3\rangle\}$ are

$$[H_\Omega]^{\mathcal{S}} = -\hbar\begin{pmatrix}\Omega_{11} & \Omega_{21}^* & \Omega_{31}^*\\ \Omega_{21} & \Omega_{22} & \Omega_{32}^*\\ \Omega_{31} & \Omega_{32} & \Omega_{33}\end{pmatrix}, \tag{22}$$

and the representation of H_Ω in the subspace $\mathcal{A}$ spanned by the antisymmetric states $\{|a_1\rangle, |a_2\rangle, |a_3\rangle\}$ is given by $[H_\Omega]^{\mathcal{A}} = -[H_\Omega]^{\mathcal{S}}$. Note that the collective ground state $|4,4\rangle$ and the states $|i,j\rangle$ ($i,j\in\{1,2,3\}$) where each atom is in an excited state are not influenced by the dipole-dipole interaction and thus not part of the expansion (21). We define $\boldsymbol{R}_z$ to be parallel to the z-axis, i.e. $\boldsymbol{R}_z = R\,\boldsymbol{e}_z$. The orientation of the separation vector $\boldsymbol{P} = \mathcal{R}_{\boldsymbol{u}}(\alpha)\boldsymbol{R}_z$ can then be adjusted at will by a suitable choice of the rotation axis $\boldsymbol{u}$ and the angle α. The explicit calculation of the coupling constants Ω_{ij} shows that the off-diagonal elements in Eq. (22) vanish if the atoms are aligned along the z-axis. It follows that the Hamiltonian

$H_\Omega(\boldsymbol{R}_z)$ is already diagonalized by the symmetric and antisymmetric states, and the eigenvalues of $[H_\Omega]^{\mathcal{S}}$ and $[H_\Omega]^{\mathcal{A}}$ are given by $\lambda_i^{\mathcal{S}} = -\hbar\Omega_{ii}(\boldsymbol{R}_z)$ and $\lambda_i^{\mathcal{A}} = \hbar\Omega_{ii}(\boldsymbol{R}_z)$, respectively. According to Eq. (12a), the Hamiltonian $H_\Omega(\boldsymbol{P})$ is the unitary transform of $H_\Omega(\boldsymbol{R}_z)$ by W. The normalized eigenstates of $H_\Omega(\boldsymbol{P})$ are thus determined by $W|s_i\rangle$ and $W|a_i\rangle$, and their eigenvalues are again $\lambda_i^{\mathcal{S}}$ and $\lambda_i^{\mathcal{A}}$, respectively. Since the orientation of $\boldsymbol{P}$ is arbitrary, the eigenvalues of $H_\Omega(\boldsymbol{P})$ depend only on the interatomic distance $|\boldsymbol{P}| = |\boldsymbol{R}_z| = R$, but not on the orientation of the separation vector.

Two additional conclusions can be drawn from Eq. (12) if the operator H_A commutes with the transformation $W = W_{\boldsymbol{u}}(\alpha)$. First, the density operator $\varrho(\boldsymbol{P})$ for $\boldsymbol{P} = \mathcal{R}_{\boldsymbol{u}}(\alpha)\boldsymbol{R}$ is then the unitary transform of $\varrho(\boldsymbol{R})$ by W, i.e. $\varrho(\boldsymbol{P}) = W\varrho(\boldsymbol{R})W^\dagger$. If the Zeeman splitting δ vanishes, H_A commutes with $W_{\boldsymbol{u}}(\alpha)$ for an arbitrary choice of the rotation axis $\boldsymbol{u}$ and angle α. We conclude that it suffices to determine the solution of the master equation (6) for only one particular geometry if $\delta = 0$. Any other solution can then be generated simply by applying the transformation $W = W_{\boldsymbol{u}}(\alpha)$ with suitable values of $\boldsymbol{u}$ and α to the solution for the particular geometry. Furthermore, the commutation relation $[H_\mathrm{A}, W] = 0$ in conjunction with Eq. (12a) implies that $H_\mathrm{A} + H_\Omega(\boldsymbol{P})$ is the unitary transform of $H_\mathrm{A} + H_\Omega(\boldsymbol{R})$ by W. If the Zeeman splitting δ vanishes, this relation holds for an arbitrary orientation of $\boldsymbol{P}$ such that the energy levels of the full system Hamiltonian $H_\mathrm{A} + H_\Omega$ do not depend on the the orientation of the separation vector. This result can be understood as follows. In the absence of a magnetic field ($\delta = 0$), there is no distinguished direction in space. Since the vacuum is isotropic in free space, one expects that the energy levels of the system are invariant under rotations of the separation vector $\boldsymbol{R}$. By contrast, the eigenvalues of the full system Hamiltonian $H_\mathrm{A} + H_\Omega$ depend on the relative position of the atoms if the Zeeman splitting δ is non-zero.

Finally, we emphasize that the result in Eq. (12) and all its implications cannot be recovered if any of the Zeeman sublevels of the P_1 triplet are neglected. In this case, the unitary operator W does not exist since it is impossible to define an angular momentum or vector operator in a state space where magnetic sublevels have been removed artificially. It follows that all Zeeman sublevels generally have to be taken into account. In order to give a more intuitive explanation of this result, we return to the matrix representation of $[H_\Omega]^{\mathcal{S}}$ in Eq. (22). The diagonal elements proportional to Ω_{ii} account for the coherent interaction between a dipole of one of the atoms and the corresponding dipole of the other atom. By contrast, the off-diagonal terms proportional to Ω_{ij} with $i \neq j$ arise from the vacuum-mediated interaction between orthogonal dipoles of different atoms [19, 20]. It is the presence of these terms that renders the simplification of the atomic level scheme impossible since they couple an excited state $|i\rangle$ of one atom to a different excited state $|j\rangle$ ($i \neq j$) of the other atom. The same applies to the collective decay rates Γ_{ij} appearing in $\mathcal{L}_\gamma\varrho$. Conversely, one can conjecture that few-level approximations are justified for particular geometrical setups where some or all of the cross-coupling terms vanish. For example, we mentioned earlier that all cross-coupling terms vanish if the atoms are aligned along the z-axis. In this case, the $S_0 \leftrightarrow P_1$ transition may be reduced to a two-level system, formed by an arbitrary sublevel of the P_1 triplet and the ground state S_0. As a second example, we assume the atoms to be aligned in the x-y-plane. Then the terms Ω_{21}, Γ_{21} and Ω_{32}, Γ_{32} vanish. In effect, the excited state $|2\rangle$ may be disregarded such that the atomic level scheme simplifies to a V-system formed by the states $|1\rangle$ and $|3\rangle$ of the P_1 multiplet and the ground state S_0.

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$-i[H_A, \varrho]/\hbar$ of the master equation is not approximated.